Review of the manuscript entitled "Relationships between particles, cloud condensation nuclei and cloud droplet activation during the third Pallas Cloud Experiment" by Martin Gysel, submitted to ACP by Anttila et al.

General comment

This study presenting results from the third Pallas Cloud Experiment (PaCE-3) builds up on previous studies (PaCE-1 and PaCE-2) by Lihavainen et al. (2008) and Anttila et al. (2009) who presented measurements at the Pallas station to indirectly determine the cloud droplet number concentration (CDNC) as well as a computational scheme which allows to infer the cloud peak supersaturation from the measured CDNC, particle number size distribution and aerosol hygroscopicity data. The data analyses presented in this study go beyond the previous papers in so far as the parcel model is used for sensitivity analyses regarding the relative importance of considering the variability of number size distribution, aerosol hygroscopicity and updraft velocity on the resulting CDNC.

Some results of this study such as the fact that, for the meteorological conditions and aerosol loadings encountered at the Pallas station, the CDNC is mainly determined by the available CCN while updraft velocity has only a minor influence are of importance.

The experiments and modeling seem to be done with care and state-of-the-art methods and the manuscript is overall well written. However, several issues regarding the interpretation and discussion of the results, which need to be addressed before final publication of this study, are provided in the following.

Major comments

1) p. 13706, l. 1-4: A positive correlation between D50 and CCN(0.4%) is reported here and it is speculated that "This can be feasibly interpreted so that larger numbers of CCN led to decreased activation efficiency due to competition between particles for water vapor during the cloud formation." However, this speculation is in contradiction to the statement made on p. 13703, l.1-3: "No visible correlation can be seen between D50 and CN(>100 nm) which suggests that the observed variation in D50 was not driven by the aerosol number concentrations." Furthermore, the authors essentially acknowledge themselves on p.13706, l. 4-9 that the correlation between D50 and CCN(0.4%) is most likely just a random result caused by poor statistics (just three cloud events): "Interestingly, only weak correlation was found between D50 and CN(>100 nm) while CDNC and CN(>100 nm) were correlated to a significant degree (Sect. 4.1). When limiting the comparison between D50 and CN(>100 nm) for the time periods during which the CCN measurements are available, however, a positive correlation with the coefficient of determination being 0.78 is seen (not illustrated here)." In conclusion, the question whether or not the CCN number concentration has an effect on the D50 has to be addressed in a more stringent way. A good estimate of CCN(0.4%) can be obtained for the whole data set by calculating the critical diameter for CCN activation at SS=0.4% from the time-resolved hygroscopic growth factor measurements (the GF values given in Table 1 indicate that kappa varies between 0.064 and 1.61, resulting in critical activation

diameters of 81-110 nm, assuming T=25 °C) and then integrate the number size distributions above this time-resolved critical diameter. If there is no correlation between this estimate of the CCN number concentration and the D50 for the whole data set, then the correlation observed for the limited data set is likely just random. However, statistics would remain limited and therefore the parcel model should be used to systematically quantify whether water vapor competition effects are to be expected in the observed range of CCN number concentrations. Further down in the manuscript (p. 13710, l. 1-4) comes a statement that the model indicates effects of the CCN number concentration differences on the resulting supersaturation. The discussion of the influence of CCN number concentration based on different analyses should be connected and consistent.

2) Table 1: The kappa values reported in Table 1 are inconsistent (too small) with the GF values. It seems that a water activity of 0.9 was used to infer the kappa values rather than an RH of 90% (the corresponding water activity will be lower due to the Kelvin effect). For example the kappa value corresponding to GF=1.24 at RH=90% and T=20 °C is ~0.118. The kappa values should be corrected. The difference in kappa values is small, however, it will also systematically affect the hygroscopicity-CCN closure presented in Section 5.

3) Sect. 5: This section is considered to be rather weak for several reasons:

a) The accuracy of the SMPS is a crucial factor for the result of hygroscopicity CCN closure studies. Was the SMPS compared against an independent measurement? E.g. comparison of the integrated CN number concentration measured by the SMPS compared with a total CN measurement made by a CPC, of course restricted to time periods when only few particles were present in the lower cut-off range of the SMPS (i.e. no nucleation mode present).

b) Why is the CCN closure only done for the cloud periods? Instead it should be done for the whole measurement campaign in order to get a more representative picture. Or is it expected that the closure would be systematically different between cloud free and cloudy periods?

c) It is speculated that the observed closure bias might be caused by the dependence of kappa on water activity. This might indeed play a role for low SS with high critical diameter (particularly for such low kappa values as reported in this study). However, the biggest closure bias is observed at the highest SS, where the CCN number concentration becomes very insensitive to the kappa value (see e.g. Fig. 8 in Juranyi et al. (2010) for a detailed sensitivity analysis. The key question here is whether the 20% bias are within or outside the experimental uncertainty of the SMPS data.

d) The hygroscopicity-CCN closure results are not at all put in the context of existing literature, e.g. Kammermann et al. (2010) and references therein, Jurányi et al. (2010), Sihto et al. (2012) (closure done for the critical diameter, which makes the comparison a little more difficult) and certainly further recent publications.

e) The HTDMA derived kappa values are not correctly calculated, which affects the closure result (see previous comment made to Table 1).

4) p. 13707, l. 17-18: "The model uses also the Koehler theory which was found to explain the CCN activity of the observed particle quite accurately (Sect. 5)" It is not directly obvious why the hygroscopicity-CCN closure should play a role for the box model. Sect. 3.3 gives the answer: The hygroscopicity of the aerosol was initialized using the HTDMA data. Would it be possible to initialize the aerosol hygroscopicity directly based on the CCN+SMPS data? This approach might result in less accurate description of the hygroscopic growth at subsaturated RH before activation in favor of a more accurate description of the activation behavior of the particles at supersaturated RH, which is of course more important for the cloud simulations. The mixing state information would be lost or could be taken from the HTDMA (taking just the GSD translated to kappa-variability for the HTDMA). Anyway, the influence of the closure bias on the uncertainty of the resulting updraft velocities / cloud peak supersaturations should be quantified (together with other uncertainties such as that of the SMPS number concentration).

5) p. 13708, l. 11 - p. 13709, l. 2: Good reproduction of the measured activation curves by the model is obtained for cloud periods D and E, while substantial differences of the shape and D50 are reported for cloud periods A-C. The authors state that the reason for this remains unknown. One obvious difference in the experimental results is that 100% activation is reached during cloud events D and E, while a stable activation plateau at around 75-90% is reached at diameters >150 nm for cloud events A-C (Why are the activation curves only shown up to 250 nm while the measurement was done up to 500 nm?). Such activation plateaus below unity can be caused by either the presence of an externally mixed non- or less hygroscopic mode or by cloud processes. Mixing state effects are excluded by the authors, leaving cloud processes as the cause of the activation plateaus below unity. Such activation plateaus can either be caused by entrainment or by evaporation of cloud droplets due to the Bergeron-Wegener-Findeisen process in mixed phase clouds. Such activation plateaus have previously been reported by e.g. Henning et al. (2004) and Verheggen et al. (2007). The parcel model has to be adapted in cases with a non-unity activation plateau (cloud periods A-C) using reasonable assumptions to simulate entrainment or droplet evaporation and using the plateau level as a constraint for the degree of these processes, such that the activation plateau is eventually reproduced by the parcel model. Will the modified simulations change the estimated updraft velocities and peak supersaturations substantially?

6) Fig. 6: The shape of the measured activation curve is surprisingly well reproduced by the parcel model. The width of the activation curve should be determined by the degree of external mixing of the aerosol as well as the inhomogeneity of the updraft velocity at cloud base. To my understanding the hygroscopic mixing state was considered with the parcel model, while no updraft velocity fluctuations were simulated. Does the agreement between experiment and model imply that the observed width of the activation curve can fully be explained by the external mixing of the aerosol, while updraft velocity fluctuations only had a marginal broadening effect on the activation curve?

7) p. 13710, l. 21 - p. 13711, l. 20: It is observed that the CCNC measures higher CCN number concentrations than the observed CDNC at equal supersaturation. Some reasons for this discrepancy are appropriately discussed. However, there are further caveats:

a) Any observed difference should be put in the context of experimental uncertainties. This is done for the kappa uncertainty. However, for example a difference of 15% in number concentration would fall within experimental uncertainty if the SMPS was undercounting by 10% and the CCNC was over counting by 5%.

b) There is a conceptual problem in the way how the CCNC measurements and the CDNC number concentrations are compared. The Kelvin effect introduces considerable temperature dependence in the CCN activation behavior, even if the kappa value at activation is assumed to be independent of temperature. To give an example, prescribing a kappa value of 0.1 and a supersaturation of 0.4% results in critical activation diameters of 94.8 nm and 105.6 nm at temperatures of 25 °C and 5 °C, respectively. This activation diameter difference roughly corresponds, according to Figs. 8 and 10, to a change in maximal cloud supersaturation from 0.4% to 0.3%. Consequently comparing CCN number concentration measurements made at ~25 °C in the CCNC column directly with CDNC number concentrations of a cloud with a temperature of 5 °C at cloud base is like comparing apples and oranges. However, there is a way to get around this issue: In the first step the CCN number concentrations and SMPS size distributions are used to infer a critical activation diameter corresponding to the supersaturation and temperature in the CCNC. These values allow it then to calculate a kappa value of the aerosol at the activation diameter. This kappa value is in a second step used to calculate the corresponding activation diameter at the cloud base temperature of the parcel model (assuming that kappa is temperature independent and only weakly dependent on particle size). Integrating the CN number size distribution above this corrected activation diameter then provides a corrected CCN number concentration measurement recalculated to cloud base temperature, which can be directly compared with the CDNC values.

8) p. 13714, l. 19-23: The authors conclude with: "It should be noted, however, that the current study is based on a rather short intensive campaign where the range of atmospheric conditions encountered was limited. Therefore long term simultaneous measurements of aerosols, CCN and cloud droplet activation are desirable to investigate how the results obtained here compare to larger data sets containing results from different seasons and air mass types." I agree that only limited data are available for the variability of updraft velocity and there is not much that can be done about this without massive additional experimental effort. However, I am sure that much more data are available about the variability and mean values of aerosol number size distribution and aerosol hygroscopicity, possibly from previous campaigns. These measurements don't have to be acquired concurrently with CDNC measurements (a reasonable range of updraft velocities can just be taken from this study) and possibly not even within clouds (unless aerosol properties were substantially different during cloud periods than outside cloud periods). Larger and more representative data sets could therefore be considered for the sensitivity analyses made in Sect. 6.2.

Minor comments

9) p. 13693, l. 28-29: Further closure/sensitivity studies of this nature are e.g: Kammermann et al., 2010 and Jurányi et al., 2010

10) p. 13696, l. 16-20: Average meteorological conditions would be at least as interesting for the cloud periods only, as the majority of data is reported for cloud periods.

11) p. 13697, l. 22: Briefly explain how the CCN was calibrated?

12) p. 13697, l. 25: Five minutes may not be enough time for the CCNC to fully stabilize when the applied SS is changed from 1% to 0.2%. The DMT CCN indicates "stabilized" temperatures much earlier than this is truly the case. Please confirm that stabilization of the CCN instrument was carefully assured.

13) p. 13698, l.3-5:

a) How was the ambient RH measured? Measurement of RH close to 100% RH may be difficult depending on the method. On the other hand, using a dew point measurement behind the total inlet and a reliable ambient temperature measurement can provide a reliable measurement of the total cloud water content.

b) How was the visibility determined, particularly during night time?

14) p. 13699, l. 20: Here it is described how the interpolation in size space is handled. What about the interpolation in time? Is the size dependence fitted for each full cycle of HTDMA data?

15) p. 13700, l. 12-15: Fitting the measured growth factor distribution with a lognormal function before determining the activated fraction is an unnecessary approximation step which potentially introduces errors. The activated fraction can directly be calculated from the measured growth factor distribution without any approximation (except for interpolation in size and time space of course), as shown in detail by Kammermann et al., 2010. The errors introduced by the lognormal approximation are likely very small, however, it is often simpler to make an accurate calculation without approximations rather than arguing that certain approximations don't cause substantial bias.

16) p. 13701, l. 14: How sensitive are the model results to the assumed mass accommodation coefficient of water? Is it possible to give some kind of a limit below/above which the model results are sensitive/insensitive to changes in the mass accommodation coefficient?

17) p. 13702, l. 19: Please explain in the experimental section that the CDNC is indirectly obtained from the difference of the particle number concentrations behind the total and interstitial aerosol inlets. Henning et al. (2002) nicely showed for liquid clouds that this indirect approach provides reliable values of the CDNC.

18) p. 13702, l. 24: It would be nice to compare the D50 values observed in this study with D50 values from other sites (e.g. Henning et al., 2002).

19) p. 13703, l. 25: I guess that the GF values are interpolated in diameter space and averaged over the duration of the cloud events.

20) p. 13704, l. 16: It might be worth repeating here that the Pallas site has a strong boreal influence.

21) p. 13705, l. 10: It would not be out of scope to put the observed kappa values briefly into the context of results reported from the boreal site Hyytiälä (Cerully et al., 2011; Sihto et al., 2012).

22) p. 13712, l. 20-21: Sensitivity analysis using the parcel model: "...For the cloud events B and C, however, the modeled values of CDNC showed largest sensitivity to the particle hygroscopicity..." This statement is potentially misleading as it seems to imply that cloud events B and C are more sensitive to aerosol hygroscopicity than to aerosol size distribution. However, this result possibly just reflects that for these two cloud events the CN size distribution in the CCN active size range was close to the averaged size distribution, while the aerosol hygroscopicity was clearly lower than the averaged aerosol hygroscopicity.

23) Fig. 5: It should be emphasized in the figure caption that this excellent agreement is forced by varying the updraft velocity in the model until agreement is achieved. This figure is all about showing that the chosen resolution of 0.05 m/s in updraft velocity steps is sufficient to reproduce the CDNC reasonably close. Actually, iteratively approaching the best fit updraft velocity with e.g. a simple bisection method rather than using a fixed updraft velocity grid would have provided perfect agreement between experiment and model with fewer model runs, thereby making this "verification"-figure obsolete.

24) Fig. 10: The data points in this figure could be colored by e.g. the CDNC or CN(>100 nm) in order to see whether the deviations from the fit line are related to the availability of CCN.

Technical corrections:

25) p. 13699, l. 19-20: Suggestion: "Accordingly the experimentally determined size dependence of the hygroscopicity parameters ..."

26) p. 13703, l. 9: The reference should be to Fig. 2 instead of Fig. 1.

27) p. 13704, l. 19: Add the Jaatinen et al. (2012, in preparation) paper to the reference list (with the tentative title and author list).

28) p. 13705, l. 15: Do you refer to Fig. 2 or Fig. 3?

29) p. 13713, l. 27: Should read: "The second part of the modeling ..."

30) Table 1: Reporting GF values without specifying the corresponding RH is useless.

References:

Cerully, K. M., Raatikainen, T., Lance, S., Tkacik, D., Tiitta, P., Petäjä, T., Ehn, M., Kulmala, M., Worsnop, D. R., Laaksonen, A., Smith, J. N., and Nenes, A.: Aerosol hygroscopicity and CCN activation kinetics in a boreal forest environment during the 2007 EUCAARI campaign. Atmos. Chem. Phys., 11, 12369-12386, doi:10.5194/acp-11-12369-2011, 2011.

Henning, S., Weingartner, E., Schmidt, S., Wendisch, M., Gäggeler, H. W., and Baltensperger, U.: Size-dependent aerosol activation at the high-alpine site Jungfraujoch (3580 m asl). Tellus, 54B, 82-95, 2002.

Henning, S., Bojinski, S., Diehl, K., Ghan, S., Nyeki, S., Weingartner, E., Wurzler, S., and Baltensperger, U.: Aerosol partitioning in natural mixed-phase clouds. Geophys. Res. Lett., 31, L06101, doi:10.1029/2003GL019025, 2004.

Jurányi, Z., Gysel, M., Weingartner, E., DeCarlo, P. F., Kammermann, L., and Baltensperger, U.: Measured and modelled cloud condensation nuclei number concentration at the high alpine site Jungfraujoch. Atmos. Chem. Phys., 10, 7891-7906, doi:10.5194/acp-10-7891-2010, 2010.

Kammermann, L., Gysel, M., Weingartner, E., Herich, H., Cziczo, D. J., Holst, T., Svenningsson, B., Arneth, A., and Baltensperger, U.: Subarctic atmospheric aerosol composition: 3. Measured and modeled properties of cloud condensation nuclei. J. Geophys. Res., 115, D04202, doi:10.1029/2009JD012447, 2010.

Sihto, S.-L., Mikkilä, J., Vanhanen, J., Ehn, M., Liao, L., Lehtipalo, K., Aalto, P. P., Duplissy, J., Petäjä, T., Kerminen, V. M., Boy, M., and Kulmala, M.: Seasonal variation of CCN concentrations and aerosol activation properties in boreal forest. Atmos. Chem. Phys., 11, 13269-13285, doi:10.5194/acp-11-13269-2011, 2012.

Verheggen, B., Cozic, J., Weingartner, E., Bower, K., Mertes, S., Connolly, P., Gallagher, M., Flynn, M., Choularton, T., and Baltensperger, U.: Aerosol partitioning between the interstitial and the condensed phase in mixed-phase clouds. J. Geophys. Res., 112, D23202, doi:10.1029/2007JD008714, 2007.